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Activation product interpretation of structural material for fast critical assemblies

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ABSTRACT

This work evaluates whether activation products in stainless steel components can be used to infer initial conditions (i.e. alloy composition, position within the fluence, etc.) and the associated uncertainties in a fast neutron irradiation. Activation measurements of stainless steel and elemental components were obtained through a Flattop critical assembly irradiation experiment. A forward model was developed, utilizing MCNP6.1 and FISPACT ii, to estimate activation product concentrations from several input parameters: alloy composition, source specifications, and foil positioning. While some of the calculated activation product concentrations agreed fairly well with measurements, some isotopics (i.e. ^{57}Co) had large discrepancies. It became clear that uncertainties in both measurements and nuclear data drove this divergence for select isotopics, and the quantification of this impact was needed. By treating both experimental measurement and nuclear data uncertainty in a Bayesian framework, it was possible to infer unknowns from given activation ratios. The developed approach showed promise in constraining mass parameters, but degeneracy existed when considering position within the neutron flux. Higher threshold reactions could potentially break the degeneracy, such as ^{57}Co, but uncertainty estimation needs to be better resolved for those isotopes.

1. Introduction

Neutron activation measurements have been utilized for a variety of problems (i.e. spectra unfolding and Neutron Activation Analysis), typically under the conditions of minimal unknowns and optimized experiments (Stankunas, 2015). However, when considering structural material irradiation under unknown conditions, uncertainty propagation can strain or break the analysis. This work evaluates whether activation products (APs) in stainless steel components can be used to infer initial conditions (i.e. alloy composition, positioning within the fluence, etc.) in a fast neutron fluence. To address this question, activation measurements of stainless steel were obtained through funded work by the Office of Defense Nuclear Nonproliferation Research and Development within the U.S. Department of Energy's National Nuclear Security Administration. The measurements were from a 2016 Flattop critical assembly irradiation coordinated through Lawrence Livermore National Laboratory (Roberts and Bandong) under controlled conditions. By utilizing measurements with constrained unknowns, a baseline for uncertainty estimates in the nuclear data could ideally be established (Otuka, 2017). The initial approach was to take the known conditions for the irradiation (i.e. foil composition, placement, and irradiation time) and model the activation product concentrations from the nuclear data. A combination of codes, MCNP6.1 (Team, 2003) (used to calculate the neutron flux) and FISPACT (Sublet, 2017) (used to solve the inventory calculations), were used as a forward model to predict activation product observables. The quality of this fit would, in principle, inform us about the state of our ability to interpret AP data in a controlled environment. Additionally, variabilities in these interpretations would be estimated (i.e. variability of a particular reaction to foil placement), establishing limitations in both the measurements and model predictions.

The first step for interpreting alloy composition and placement was to identify specific APs that were particularly sensitive to these characteristics. The reactions sought after consisted of both threshold (n,p) and non-threshold (n,γ) reactions, due to high sensitivities in multiple energy regions (spectral edge and down-scattering). By utilizing multiple APs, the measurements could be normalized to a non-threshold reaction, ^{58}Fe(n,γ)^{59}Fe, removing correlated unknowns such as the flux magnitude. It was determined early in the computational analysis that while some selected (signature) isotopic predictions were excellent

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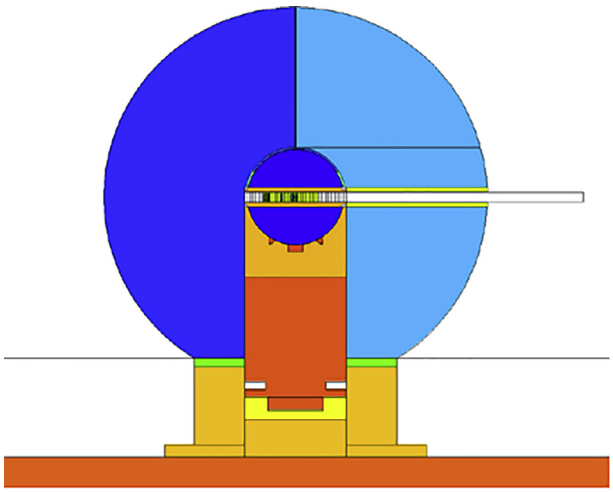

Fig. 1. Detailed MCNP schematic of Flattop with a HEU core.

compared to the measurements, others had a poor fit. It became clear that while uncertainties in the nuclear data had some impact, experimental uncertainties (foil composition and placement) also influenced the results. In order to quantify the impact of multiple uncertainties associated with the signature isotopes, we developed a Bayesian methodology to test the sensitivity of alloy/placement predictions.

2. Methodology

One goal of this work was to evaluate whether a gamma measurement could differentiate various grades of stainless steel, mainly the nickel and chromium concentrations of an unknown alloy. Nickel stabilizes the austenitic structure of the steel and increases the high-temperature strength, and chromium forms a passive surface to increase resistance to corrosion (Barnard, 2017). By inferring the concentration of those elements in an alloy, some conclusions can be drawn for the potential application.

3. Flattop-25 measurement/computational methods

In order to evaluate the capability, activation measurements of both stainless steel and stainless steel component (i.e. pure Cr, Ni, Fe) foils were obtained through a 2016 Flattop (^{235}U core) critical assembly irradiation. The core includes two highly enriched uranium (~93.2 wt%) metal hemispheres with a total mass of 16.133 kg, surrounded by a normal uranium reflector when assembled (NEA, International, 2016). Three foil packets (0.318 cm width), consisting of multiple stainless steel and elemental components, were placed in the center of the core traverse. Gold foils were placed on the outer edges and in the center of the foil packets to monitor fluence discrepancies along the experimental package. By utilizing witness foils, the effects of the foils on the fluence (i.e. self-absorption) could be monitored. The analysis of the ^{198}Au/^{196}Au ratio between gold foils showed negligible variation within measurement uncertainty, and impact of self-absorption on the fluence was thought to be minimal.

The foils were irradiated over 0.883 h and allowed to cool for 3–15 days (depending on the sample) before being counted on a suite of ORTEC GEM series p-type coaxial HPGe detectors. Due to the large sample set and decay considerations, a suite of detectors were used for counting. To minimize bias when comparing measurements from differing detectors, samples were counted on a minimum of three different detectors to produce multiple independent measurements (i.e. utilizing different core efficiency information). The relative efficiencies of the HPGe detectors ranged from 12 to 37%, utilizing a variety of NIST-traceable point sources covering the energy region of 59–2000 keV (^{241}Am, ^{57}Co, ^{137}Cs, etc.) (Goodell, 2018). The uncertainties on NIST sources varied, but are typically 3% total uncertainty, which is factored into the statistical uncertainty of the analysis. Depending on the foil and photo-peaks under consideration, counting times and geometries were optimized for desired statistical uncertainty of the measurements. The highest gamma yields for the given irradiation (from

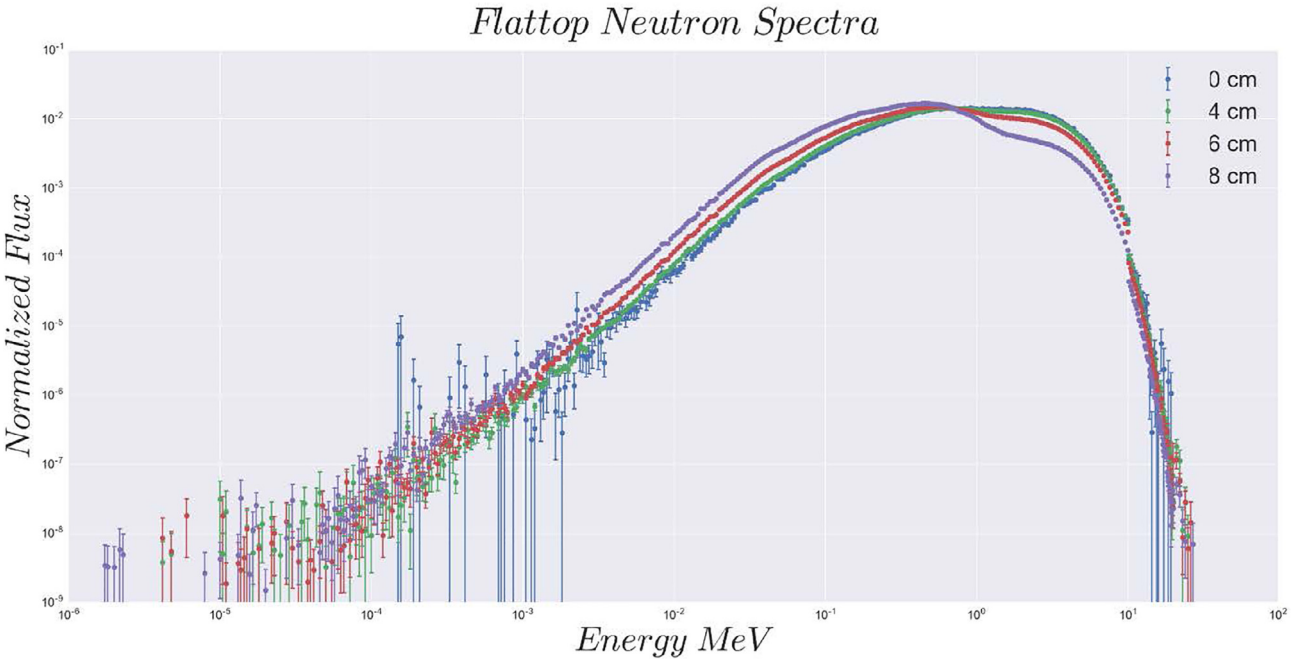

Fig. 2. Calculated neutron spectra of various foil positions within Flattop (uranium core).